Correlated Polarons in Dissimilar Perovskite Manganites

Broad peaks attributed to the presence of correlated polarons are observed in the paramagnetic insulating phases of $La_{0.7}Ca_{0.3}MnO_3$ and $Pr_{0.7}Ca_{0.3}MnO_3$. In the two samples, which exhibit strikingly different low-temperature phases (ferromagnetic metallic, and charge and orbitally ordered insulating, respectively), the correlated polarons are found to behave similarly. Specifically, the broad peaks occur at identical wavevectors, exhibit temperature-independent correlation lengths of 1-2 lattice constants, and gradually increase in intensity with decreasing temperature. These surprising results indicate the robust nature of the correlated polarons to variations in the lattice degree of freedom.

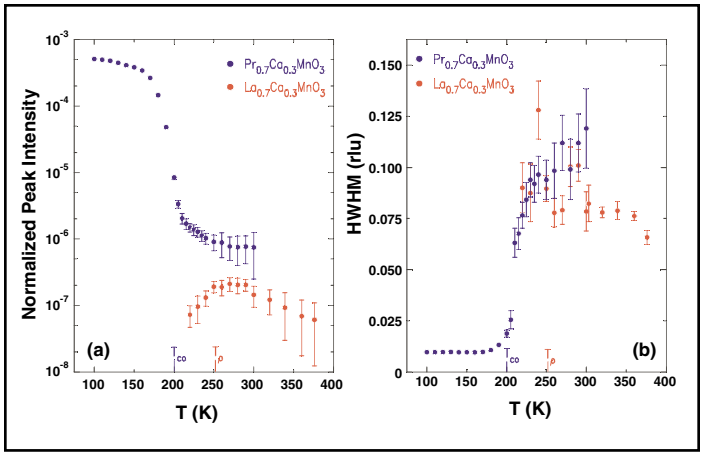

FIG. 1. Peak intensity (a) and HWHM (b) of the (½00)/(0½0)-type scattering. The intensities are normalized to the (220) Bragg peak intensity at 220K ($La_{0.7}Ca_{0.3}MnO_3$) and 100K ($Pr_{0.7}Ca_{0.3}MnO_3$). The intensity data for T ≥300K of $La_{0.7}Ca_{0.3}MnO_3$ were scaled to match the low-temperature data at 300K. Note that the vertical lines labeled T_{co} and T_ρ indicate the charge and orbital-order transition and metal-insulator transition temperatures in $Pr_{0.7}Ca_{0.3}MnO_3$ and $La_{0.7}Ca_{0.3}MnO_3$, respectively.

One of the many interesting properties exhibited by some transition metal oxides is an extraordinarily large change in resistivity upon the application of a magnetic field—i.e., the so-called colossal magnetoresistance (CMR) effect. As was first pointed out by Millis et al. [1], electron-phonon coupling is very important for explaining the behavior of CMR materials and has therefore been the focus of much recent experimental work on these materials [2]. In the CMR manganites, strong electron-phonon coupling results in the formation of charge carriers with associated lattice distortions, or polarons. These polarons can be probed using scattering techniques, with which both single polarons and correlated polarons can be investigated. In what follows, we describe an x-ray scattering study of correlated polarons in dissimilar perovskite manganites [3], which differ only with respect to their trivalent cation species. The two samples are $La_{0.7}Ca_{0.3}MnO_3$ and $Pr_{0.7}Ca_{0.3}MnO_3$.

$La_{0.7}Ca_{0.3}MnO_3$ and $Pr_{0.7}Ca_{0.3}MnO_3$ are paramagnetic insulators at room temperature, and exhibit completely different low-temperature phases. While $La_{0.7}Ca_{0.3}MnO_3$ is a ferromagnetic metal below 252K, $Pr_{0.7}Ca_{0.3}MnO_3$ is a charge and orbitally ordered insulator below ~200K, and orders antiferromagnetically at ~130K. The origin of this difference in the low-temperature phases is believed to be the ~3% decrease in cation radius from La to Pr, which also increases the relative strength of the electron-phonon coupling. Our motivation was to study the effects of this difference on the paramagnetic insulating phases of the two samples.

The measurements were carried out at beam-

line X22C at the National Synchrotron Light Source (NSLS) and at CMC-CAT beamline 9-ID at the Advanced Photon Source (APS). The low-temperature work was performed using a closed-cycle refrigerator. For the high-temperature measurements of $La_{0.7}Ca_{0.3}MnO_3$, the sample was placed in a furnace in an air atmosphere. Both samples were determined to be fully twinned, with (110)/(002)-oriented surface normals (in orthorhombic, *Pbnm* notation). For simplicity, the peaks described here are referenced assuming a (110) surface normal.

In both $La_{0.7}Ca_{0.3}MnO_3$ and $Pr_{0.7}Ca_{0.3}MnO_3$, broad peaks attributed to the presence of correlated polarons are observed at $(\frac{1}{2}00)/(0\frac{1}{2}0)$-type wave vectors. In Fig. 1(a), the temperature dependences of the fitted peak intensities are shown. The peak intensities in both samples are observed to increase gradually with decreasing temperature and then diverge at the respective transition temperatures. In Fig. 1(b), the temperature dependences of the fitted HWHM values are displayed. Again, the high-temperature behaviors are found to be similar in the two samples. The correlation length—obtained using $\xi = a/(2\pi \times \text{HWHM})$, where a is the lattice constant—is independent of temperature with a magnitude of 1 to 2 lattice constants.

The wave vector and the magnitude and temperature dependence of the correlation length of the broad peaks suggest that the correlated polarons arise from similar composite objects in the two samples [4]. A proposed structure for these objects is shown in Fig. 2. It consists of two Mn^{4+} ions situated between orbitally ordered Mn^{3+} ions and resembles an orbital order domain of the CE-type structure. We note that, while this proposed structure is consistent with all of our results, it is not unique and alternative structures can also be constructed.

In conclusion, we studied two CMR manganites that differ only with respect to their trivalent cation species. In sharp contrast to the sensitivity of the low-temperature phases, we find that the correlated polarons in the paramagnetic insulating phases of the two samples are robust with respect to variations in the lattice degree of freedom.

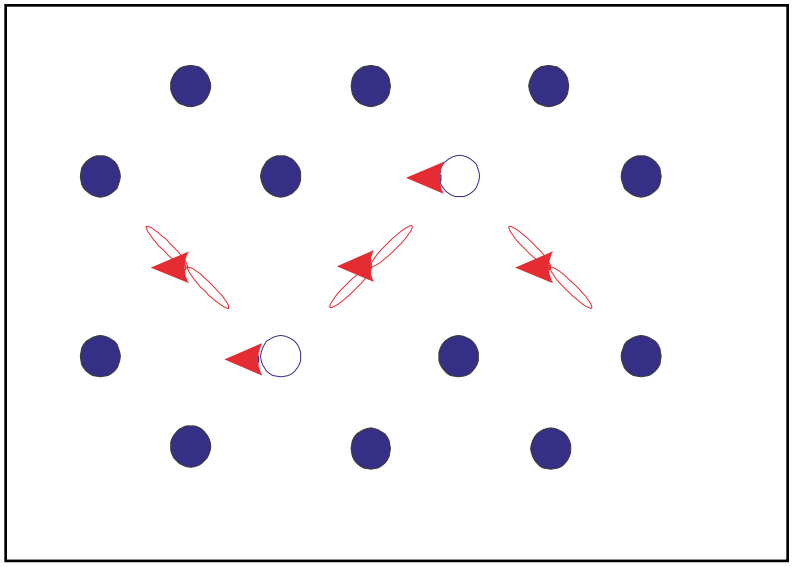

FIG. 2. Proposed in-plane structure of the correlated polarons. Open circles represent Mn^{4+} ions; elongated figure-eights represent the occupied e_g ($3d_{z^2-r^2}$) orbital of Mn^{3+} ions; closed circles represent Mn ions that, on average, have the formal valence and no net orbital order; and arrows indicate the in-plane component of the magnetic moment.

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[2] See, for example, S.J.L. Billinge et al., Phys. Rev. Lett. **77**, 715 (1996); Shengalaya et al., Phys. Rev. Lett. **77**, 5296 (1996); Shimomura et al., Phys. Rev. Lett. **83**, 4389 (1999); Vasiliu-Doloc et al., Phys. Rev. Lett. **83**, 4393 (1999).

[3] For a complete description of this study, see Nelson et al., cond-mat/0011502, Phys. Rev. B, to be published.

[4] Note that Kiryukhin et al., in preparation, report similar results in another x = 0.3 perovskite manganite, $Nd_{0.7}Sr_{0.3}MnO_3$.

C. S. Nelson,[1] M. V. Zimmermann,[2] Y. J. Kim,[1] J. P. Hill,[1] D. Gibbs,[1] V. Kiryukhin,[3] T. Y. Koo,[3] S-W. Cheong,[3,4] D. Casa,[5] B. Keimer,[6] Y. Tomioka,[7] Y. Tokura,[7,8] T. Gog,[5] C. T. Venkataraman[5]

[1] *Department of Physics, Brookhaven National Laboratory, Upton, NY, U.S.A.*
[2] *HASYLAB at DESY, Hamburg, Germany*
[3] *Department of Physics and Astronomy, Rutgers University, Piscataway, NJ, U.S.A.*
[4] *Bell Laboratories, Lucent Technologies, Murray Hill, NJ, U.S.A.*
[5] *CMC-CAT, Advanced Photon Source, Argonne National Laboratory, Argonne, IL, U.S.A.*
[6] *Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany*
[7] *Joint Research Center for Atom Technology (JRCAT), Tsukuba, Japan*
[8] *Department of Applied Physics, University of Tokyo, Tokyo, Japan*